

EXHIBIT 12



Evaluation of PCB sources and releases for identifying priorities to reduce PCBs in Washington State (USA)

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Abstract Polychlorinated biphenyls (PCBs) are ubiquitously distributed in the environment and produce multiple adverse effects in humans and wildlife. As a result, the purpose of our study was to characterize PCB sources in anthropogenic materials and releases to the environment in Washington State (USA) in order to formulate recommendations to reduce PCB exposures. Methods included review of relevant publications (e.g., open literature, industry studies and reports, federal and state government databases), scaling of PCB sources from national or county estimates to state estimates, and communication with industry associations and private and public utilities. Recognizing high associated uncertainty due to incomplete data, we strived to provide central tendency estimates for PCB sources. In terms of mass (high to low), PCB sources include lamp ballasts, caulk, small capacitors, large capacitors, and transformers. For perspective, these sources (200,000–500,000 kg) overwhelm PCBs estimated to reside in the Puget Sound ecosystem (1500 kg). Annual releases of PCBs to the environment (high to low) are attributed to lamp ballasts (400–1500 kg), inadvertent generation by industrial processes (900 kg), caulk (160 kg), small capacitors (3–150 kg), large

capacitors (10–80 kg), pigments and dyes (0.02–31 kg), and transformers (<2 kg). Recommendations to characterize the extent of PCB distribution and decrease exposures include assessment of PCBs in buildings (e.g., schools) and replacement of these materials, development of Best Management Practices (BMPs) to contain PCBs, reduction of inadvertent generation of PCBs in consumer products, expansion of environmental monitoring and public education, and research to identify specific PCB congener profiles in human tissues.

Keywords Polychlorinated biphenyls · PCBs · Caulk · Inadvertent generation · Washington State · Sources

Introduction

Polychlorinated biphenyls (PCBs) were widely used in the USA before they were banned in 1979 under the Toxics Substances Control Act (TSCA). From 1929 to 1979, about 600,000 metric tons of PCBs were commercially manufactured in the USA (EPA 1997). About 75 % of PCBs produced before 1979 were used in transformers and capacitors, including small capacitors in lamp ballasts and appliances. The second largest pre-1979 use (about 10 %) was as plasticizers, including in caulk (EPA 1997). TSCA allowed some historical uses of PCBs to continue and set allowable levels of inadvertent production of PCBs in other products (e.g., pigments and dyes). In our study, we refer to these PCB sources (e.g., transformers, capacitors, caulk, pigments, dyes) as “anthropogenic materials.” Collectively, PCBs in anthropogenic materials comprise a relatively large source of PCBs in Washington State and are the focus of our analysis.

PCBs are also regulated under additional state and federal laws, which are sometimes inconsistent. For example, the levels of PCBs allowed from inadvertent generation under

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TSCA are many orders of magnitude larger than the levels of PCBs allowed in water under the Clean Water Act. Clean water standards will be met, only if all sources of PCBs, including inadvertent generation, are addressed.

There is a large body of toxicological and epidemiological research on the health effects of PCBs, e.g., summarized in USDHHS (2000). The International Agency for Research on Cancer (IARC) recently changed their classification of PCBs from “probable human carcinogens” to “human carcinogens” to recognize that there is now sufficient evidence that PCBs cause cancer in humans and animals (Lauby-Secretan et al. 2013). PCBs also cause noncancer adverse effects in humans, including harm to immune (Heilmann et al. 2006), neurobehavioral and development (Jacobson and Jacobson 1996; Jurewicz et al. 2013), endocrine (Hamers et al. 2011), and reproductive systems (Buck Louis et al. 2013). Similar adverse effects have been observed in wildlife, including cancer (Ylitalo et al. 2005), immune disorders (Ross et al. 1996), and reproduction and development endpoints (Bursian et al. 2013).

The general US population has detectable levels of PCBs in their blood (CDC 2009). Although body burdens of PCBs have declined by more than 80 % since the 1980s, the levels are no longer decreasing. Levels of PCBs in the 2003–2004 National Health and Nutritional Examination Study (NHANES) are roughly similar to the previous two NHANES survey periods (CDC 2009). Similarly, levels in wildlife (Krahn et al. 2007) and the environment (Davis et al. 2007) have also decreased dramatically since the 1980s. However, the decrease has plateaued, and PCBs are still widespread. Wildlife is exposed to PCBs in diet, along with PCBs in water, soil, and sediments (Eisler 1986). PCBs accumulate in fatty tissues, including in animals eaten for food. PCBs in food are the most significant source of exposure for most humans (USDHHS 2000), and we are particularly concerned with levels of PCBs in fish we eat (e.g., Delistraty 2013). Humans are also exposed to PCBs in air, water, soil, and house dust.

Washington State recognizes the special concerns and challenges, involving persistent, bioaccumulative, and toxic chemicals (PBTs), such as PCBs, and has a strategy to better understand and take actions to further reduce PBTs (Ecology 2000; WAC 173-333 2006). The foundation of the strategy is the production and implementation of Chemical Action Plans (CAPs) to identify, characterize, and evaluate uses and releases of a specific PBT or a class of structurally and functionally related PBTs and recommend actions to protect human health and the environment. The Washington Departments of Ecology and Health have worked with multi-stakeholder advisory committees to complete five CAPs on methyl mercury, polybrominated diphenyl ethers (PBDEs), lead, polycyclic aromatic hydrocarbons (PAHs), and PCBs. More information

on PCBs in Washington can be found in the PCB CAP (Ecology 2015).

This study describes our evaluation of PCB sources in anthropogenic materials in Washington State, along with historical uses and releases of PCBs to the environment. We acknowledge the uncertainty in our PCB estimates, as a result of numerous data gaps. For perspective, reservoirs of PCBs in environmental media (e.g., air, soil, sediment, water, biota) are briefly addressed, as a basis with which to compare (by estimated mass) PCBs in anthropogenic materials. We conclude by providing a set of recommendations to reduce and phase out uses, releases, and exposures of PCBs in Washington State, offering new and more innovative management approaches.

Materials and methods

Note that throughout our study, units are reported as given in the cited reference. In some cases, equivalent metric units are provided.

Transformers and large capacitors

The EPA PCB Transformer Registration Database contains 252 registered PCB transformers in Washington State with more than 500 ppm PCBs, totaling 121,053 kg of PCB oil (EPA 2014). This database includes “PCB transformers” that have greater than 500 ppm PCBs and does not include “PCB contaminated” transformers with 50–500 ppm PCBs or “non-PCB” transformers with less than 50 ppm PCB. This database was compiled in 1998 but has not been updated.

As a result, we contacted 11 of 14 parties who registered transformers, accounting for 242 of 252 (96 %) transformers in the EPA database to update our estimate for PCB transformers (Online Resource 1 [supplementary material]). We also spoke to two additional utilities that did not have PCB transformers registered in the EPA database. We estimated the amount of PCBs still in PCB transformers, assuming 665 gal of fluid containing 1500 ppm PCB for each transformer, based on the sizes and PCB concentrations of PCB transformers still in use. In particular, we found no evidence that Askarel transformers (typically filled with 60–70 % PCBs) were used in Washington State. Some utilities had tested all of their equipment in use and provided information on exactly how many were >500, 50–500, and 1–50 ppm PCBs. Based on information from public and private state utilities on this equipment with lower levels of PCBs that is known to still be in use, along with older equipment that is untested and has unknown levels of PCBs, we estimated there are about 40,000 PCB contaminated (50–500 ppm) and non-PCB (1–50 ppm) transformers in the state with an average PCB concentration of about 25 ppm in 75 l of fluid per transformer.

We scaled the national US estimate of 1,293,000 large capacitors in use in 2007 from The Great Lakes Binational Toxics Strategy 2009 Biennial Report (EPA/EC 2009) to 28,160 large capacitors in Washington State, based on population size. Populations were estimated from the 2010 Census, where Washington State had a population of 6,724,543 out of a total US population of 308,747,716. We contacted public and private utilities who historically owned about 85 % of large capacitors (Panero et al. 2005) and found they had replaced large PCB capacitors more than 10–20 years ago. We estimated that 5 % of the large capacitors (~1400) remain in use, mostly in non-utilities, based on the premise that non-utilities have also removed many but not all of these large capacitors. Capacitor size varies, and they were usually filled with nearly pure PCB oil. Typical large capacitors contain 31 lb (14 kg) of PCBs (EPA 1982).

Leakage and spillage rates for transformers and large capacitors were based on a national industry study (USWAG/EEI 1982), reporting that about 2 % of all transformers and 0.77 % large capacitors had moderate leaks or spills each year. This release estimate does not account for spill response, so the actual amounts of PCBs released to the environment may vary. Indoor spills in particular are likely to be contained and cleaned up. Additional emissions from direct volatilization from equipment are likely but not estimated.

Small capacitors

We used a national estimate of 870 million small capacitors (i.e., capacitors containing less than 3 lb of PCB oil) throughout the USA in 1977 in industrial machines and small appliances (EPA 1982), scaled to Washington State (based on population) and applied annual disposal rates of 20 and 10 % to approximate a range of small capacitors still in use (EPA 1987). We estimated lamp ballasts separately, based on national estimates of 300 million by the US Army in 2001, 500 million by Missoula County in 2010, and 1 billion by EPA in 1998 (Ecology 2011b). Again, we scaled our estimate to Washington State (based on population) and applied annual disposal rates of 20 and 10 % to approximate a range of lamp ballasts still in use (EPA 1987). While small capacitors may contain 45–270 g PCB per unit, most of the remaining units are likely to be lamp ballasts, which typically contain 45–70 g PCB per unit. For the estimate, we used 57.5 g PCB/unit as an average. The assumed leakage rate is 4.2 kg/metric ton of PCBs from the 1982 study on large capacitors (EPA 1982).

Caulk

We estimated the amount of PCBs in sealants, based on the number of existing masonry commercial buildings that were built between 1945 and 1980, the average size of those buildings, and the distribution of PCB concentrations in caulk. An

earlier study of the Puget Sound Basin (Ecology 2011b) collected detailed information about buildings in Pierce and Snohomish Counties in Washington State. The estimated volume of masonry buildings, constructed from 1945 to 1980 in these two counties, was 21,941,562 m³. To estimate PCBs in caulk for the state, we scaled up the volume of masonry commercial buildings that were built between 1945 and 1980 by population, leading to an estimate of 97,702,645 m³ with 5,373,645 kg of caulk for the state. The percentage of buildings with PCBs in caulk and the PCB concentration ranges from Kohler et al. (2005) were applied to the estimated mass of PCB-containing sealants in Washington State. The annual release estimate was based on a release rate coefficient of 0.0018/year from long-term loss rates in Robson et al. (2010).

Inadvertent generation of PCBs

We estimated the amount of PCBs inadvertently generated in new products, based on information submitted to EPA and scaled to Washington State by population. EPA's 1984 final rule under TSCA on inadvertent generation (49 FR 28172 1984) includes an annual estimate of 100,000 lb (45,400 kg) of PCBs. We also reviewed manufacturer reports to EPA of inadvertently generated PCBs between 2 and 50 ppm.

For our estimate of PCBs in pigments, we scaled to Washington State by population from national estimates from Guo et al. (2014) and information submitted to EPA by the Color Pigments Manufacturers Association (CPMA) in 2010. CPMA estimated that the total annual amount of these pigments (phthalocyanine and diarylide) imported or manufactured in the USA is about 90 million lb. CPMA further estimated inadvertently generated PCBs in these pigments with an upper bound of 1.1 tons/year and a more reasonable estimate of 1000 lb/year. Because inadvertently generated PCBs in new products (e.g., pigments) are not contained within the product, we assumed all of the PCBs are eventually released into the environment.

Results

Historic uses of PCBs

Transformers and large capacitors

We estimated only 100–200 kg of PCBs remain in transformers in Washington State, with less than 2 kg released each year (Table 1). The largest historic uses of PCBs were for transformers and large capacitors which have been largely replaced. By contacting most of the parties who registered PCB transformers in Washington State with EPA in 1998,

Table 1 Summary of sources, mass estimates, and releases of PCBs in Washington State

Source	Mass estimate of PCBs	Annual releases of PCBs (kg/year)
Historic uses		
Transformers	100–200 kg	<2
Large capacitors	20 metric tons	10–80
Lamp ballasts	100–350 metric tons	400–1500
Small capacitors	1–34 metric tons	3–150
Other “closed” uses	Unknown	Unknown
Caulk	87 metric tons	160
Other “open” uses	Unknown	Unknown
Current generation		
Pigments and dyes	Unknown	0.02–31 ^a
Other inadvertent generation	Unknown	900

^a This estimate is for PCB-11, although additional congeners are present

we found that 228 were disposed of, 14 are still in use, and 10 were of unknown status (Table 2 and Online Resource 1 [supplementary material]). In addition, utilities in Washington State have been voluntarily testing transformers and disposing of transformers with PCBs greater than 2 ppm.

We estimated that about 20 metric tons of PCBs still remain in 1400 large capacitors with 10–80 kg released each year (Table 1).

Small capacitors

We estimated that there are still 12,000–586,000 small capacitors with 1–34 metric tons of PCBs and an annual release of 3–150 kg (Table 1). Small capacitors containing PCBs have been used in a number of items including motors, appliances, and light ballasts. We also estimated of 1.7–6.2 million PCB lamp ballasts with 100–350 metric tons of PCBs and 400–1500 kg released each year (Table 1).

Caulk

We estimated that 87 metric tons of PCBs are in caulk in industrial masonry buildings in Washington State with 160 kg released each year (Table 1).

Table 2 Status of PCB transformers in Washington State (see Online Resource 1 [supplementary material] for more detail)

Status	Number of transformers
Still in use	14
Disposed of	228
Unknown	10
Total	252

Current inadvertent generation of PCBs

New generation of PCBs may be as high as 900 kg/year in Washington State, with 0.02–31 kg/year due to PCB-11 in yellow pigments (Table 1). This generation rate for Washington State is scaled from a national annual estimate of 100,000 lb (45,400 kg), as described in EPA’s 1984 rule under TSCA on inadvertent generation of PCBs (49 FR 28172 1984). The 100,000 lb estimate is a consensus proposal from the Environmental Defense Fund, Natural Resources Defense Council, and Chemical Manufacturers Association (now known as the American Chemistry Council) that included all inadvertent generation of PCBs, without allocating amounts to individual processes. Products specified included paints, printing inks, agricultural chemicals, plastic materials, and detergent bars.

EPA’s 1984 rule (49 FR 28172 1984) requires manufacturers to report inadvertent generation of PCBs. There are 77 reports for inadvertently generated PCBs from 1994 to present (Table 3 and Online Resource 2 [supplementary material]). Additional reports are included in the EPA docket for related topics (e.g., requests to produce small amounts of PCBs for research purposes). Much of the information in the reports has been redacted to remove confidential business information. In general, the reports repeat federal requirements, while stating the company is in compliance but without providing specific information on the concentration of PCBs in the products or total quantity of products. None of the reports come from facilities in Washington State.

Some of the reports in Table 3 in the category of pigments and dyes list individual pigments (e.g., yellow, red, green, blue violet, and orange with Color Index (CI) numbers) or include a general description (e.g., “imported dyes”), while others do not include specific information but specify a division of the company (e.g., “Pigments Division”). Eight reports are from GE Silicones with no additional information on the products. Three reports are from three different companies, regarding vinyl chloride production. One company stated it was reporting on 740 lb of PCBs in 62,676,000 lb of chemical feedstocks, used in a vinyl chloride monomer manufacturing

Table 3 Reports to EPA on inadvertent generation of PCBs (1994–present) (see Online Resource 2 [supplementary material] for more detail)

Chemical or process	Number of reports
Pigments and dyes	53
GE silicones	8
Vinyl chloride production	3
Unique	6
Unknown	7
Total	77

facility in 1995. There are six reports from six different companies on unique compounds or processes, while seven reports were for unknown compounds or processes.

Discussion

Perspective

Current PCB levels in the environment represent both historical and ongoing loadings of PCBs in anthropogenic materials (the focus of our study), as well as PCBs contained in environmental compartments (e.g., air, soil, water, sediment, biota). In the past, work has been focused on known point sources of PCBs, including water permittees, owners of large electrical equipment, and contaminated industrial sites, but newer work has shown the importance of diffuse nonpoint sources. For example, according to recent Puget Sound, Lake Washington, and Spokane River studies, the primary pathway for PCBs to reach the aquatic environment is stormwater (Ecology 2011a, Ecology 2011c; King County 2013). PCBs do not readily dissolve in water but bind to particles. In turn, these particle-bound PCBs are transported via stormwater to sediment. Direct air deposition was estimated to be the second largest pathway, while publicly owned treatment works (POTWs) comprise a less significant pathway, contributing less than 10 % of total PCB loading. Other pathways (e.g., groundwater, ocean exchange) or unknown pathways transport additional amounts of PCBs to the aquatic environment in Washington State. Similar results on the importance of stormwater runoff (compared to permitted point sources) have been found in other jurisdictions, including San Francisco (Davis et al. 2007), New York/New Jersey Harbor (Panero et al. 2005), and the Delaware River Basin (Praipipat et al. 2013).

The mass of PCBs still in use in anthropogenic materials and yet to enter the environment is much larger than the mass of PCBs already in the environment. Estimates of PCBs still in use range from 38 to 54 % of the PCBs produced (CCME 1995; EPA 1997; Keeler et al. 1993). Tanabe (1988) estimated global PCBs by mass in terrestrial and coastal systems, allocating 0.4 % to air, 2.5 % to river and lakewater, 1.7 % each to seawater and soil, 90.9 % to sediment, and 3.0 % to biota. These estimates are consistent with those for the Puget Sound ecosystem in Washington State, where approximately 97 % (1440 kg) of the total mass of PCBs is contained in the active sediment layer (top 10 cm), <1 % (10 kg) exists in the water column, and <3 % (40 kg) resides in the biota (Ecology 2011a). These mass estimates of PCBs in the Puget Sound ecosystem are comparatively small, relative to mass estimates contained in anthropogenic materials (Table 1).

Uncertainty in PCB estimates

It is acknowledged that mass estimates and annual releases of PCBs (Table 1) necessarily have a high degree of uncertainty, resulting from incomplete data on inputs required for estimation. Data on PCB sources and concentrations in materials were obtained from published information, government reports, communication with utilities and industry associations, and simple scaling methods to derive estimates for Washington State. To the extent possible, we attempted to provide central tendency estimates of the relevant distributions to increase credibility. As such, we did not intentionally incorporate conservatism or nonconservatism into our estimates. Although not expressed in powers of ten, PCB estimates (Table 1) should be viewed as rough order of magnitude approximations with large corresponding variance. The utility of these estimates is more in a relative context for prioritization, rather than in conveying quantitative accuracy.

Numerous data gaps contribute uncertainty when evaluating PCBs in anthropogenic materials. For example, transformers with less than 500 ppm PCBs, as well as capacitors, have never been required to be tracked or reported. As a result, less is known about this equipment. In addition to transformers and capacitors, utilities have other equipment that historically contained PCBs (e.g., reclosers, switches, circuit breakers, bushings, etc). Although this equipment contained much lower amounts of PCBs, compared with transformers and capacitors (USWAG/EEI 1982), we have not attempted to estimate how much of this older PCB-containing equipment remains in use. Some utilities have been testing and removing these materials. Another data gap relates to PCB-containing lamp ballasts, particularly in schools in Washington State. There is no statewide database of school buildings that would allow us to estimate how many might have PCB lamp ballasts, based on age of construction or renovations. With respect to caulk, uncertainties relate to historical use of caulk, as well as PCB concentrations in caulk.

Caulk and other building materials

PCBs were used in caulk and joint sealants from the 1950s to the 1970s to improve sealant flexibility, increase resistance to erosion, and improve adherence to other building materials (Robson et al. 2010). While the use of PCBs in “open” (i.e., dissipative) products above 50 ppm was banned in the USA in 1979 under TSCA, materials that contain PCBs were not required to be removed. The use of PCB-containing caulk was a common practice in the 1970s, and caulk formulations changed during the late 1970s (Herrick et al. 2004). Studies on PCBs in caulk have focused on buildings built from about 1950 to 1980 in an attempt to account for existing stocks of

PCB-containing caulk. Sealants with high levels of PCBs have been found at varying levels in buildings in several studies (Herrick et al. 2004; Kohler et al. 2005; Diamond et al. 2010; Klosterhaus et al. 2014; SAIC 2011). PCBs in caulk are associated with higher levels of PCBs in indoor air and dust, as well as surrounding soils (Priha et al. 2005; Herrick et al. 2004).

As part of the Lower Duwamish Waterway (LDW) cleanup in Seattle, Washington SAIC (2011) investigated PCBs in old caulk and paint in industrial buildings constructed from 1950 to 1977. This was part of an effort to identify additional local sources of PCBs, especially since high levels of PCBs in paint, caulk, and other building materials had been found in nearby areas. SAIC detected Aroclors in 8 of 17 composite caulk samples from representative buildings with detected concentrations ranging from 3 to 920 mg/kg. The number of samples with detectable PCBs (47 %) is in agreement with a larger comprehensive study in Switzerland (Kohler et al. 2005). We did not have adequate information to include in our estimate caulk in all building uses (e.g., window sealing), residential buildings, or use in other structures (e.g., bridges and sidewalks) that contribute to environmental loading. For example, stormwater sediments in Tacoma, Washington were contaminated by PCBs in caulk from nearby buildings and sidewalks (WDOH 2011).

PCBs were used in additional building materials, including paint, and paint absorbs PCBs from other sources, e.g., caulk and light ballasts (EPA 2012). We did not have adequate information to estimate PCBs in these other building materials. Although PCBs may be transferred to the environment during renovations or destruction, certain removal practices can reduce the amount of PCBs released both to workers and the environment (Sundahl et al. 1999). EPA conducted a pilot study for addressing PCBs in building materials at six schools in New York that were scheduled for major renovation or demolition and were suspected of containing PCB sources as part of a settlement of a lawsuit (EPA 2012).

Inadvertent generation of PCBs

Inadvertent generation of PCBs is a potentially large and important source of uncontained PCBs, and the current EPA regulations are not effective. PCBs are no longer intentionally manufactured in the USA, and the manufacture, processing, and distribution in commerce of PCBs at concentrations of 50 ppm or greater are not allowed. EPA promulgated a rule under TSCA in 1984 (49 FR 28172) for inadvertent generation of PCBs that are not in “closed” (i.e., controlled) manufacturing processes. The concentration of inadvertently generated PCBs in products must have an annual average of <25 ppm, with a maximum of 50 ppm. The concentration of monochlorinated biphenyls is reduced by a factor of 50 and the concentration of dichlorinated biphenyls is reduced by a

factor of 5, due to lower environmental persistence and bioaccumulation of these PCB homologues (49 FR 28172 1984). In addition, EPA required manufacturers with processes inadvertently generating PCBs and importers of products containing inadvertently generated PCBs to report to EPA any process or import for which the PCB concentration exceeds 2 mg/kg for any resolvable PCB gas chromatographic peak.

Little is known about these processes or related products, other than studies on pigments and dyes. PCBs are known to be inadvertently generated in certain pigments and dyes, including diarylides (yellow and orange), naphtharylides (oranges and reds), phthalocyanines (blue), and basic dye complex pigments (reds, violets, blues, and greens) (Christie 2013; Hu and Hornbuckle 2010). Because PCB-11 is thought to be primarily from pigment production and not from legacy uses of Aroclors (Hu and Hornbuckle 2010; Guo et al. 2014), it is useful as an indicator of inadvertent PCB generation, especially from pigments. Hu and Hornbuckle (2010) found PCBs in azo and phthalocyanine pigments, including PCB-11 and higher chlorinated PCBs 206–209. Previously, PCB-209 was only thought to be found in ferric oxide, as a by-product of titanium dioxide production (Panero et al. 2005). The Japanese Ministry of Economy, Trade and Industry (METI), along with two related ministries, analyzed 242 organic pigments found to contain PCBs as contaminants. Among these pigments, 101 contained PCBs over 0.5 ppm (METI et al. 2013a), while four yellow pigment samples contained PCBs in the range of 59–1000 ppm (METI 2013b).

PCBs that are generated as a by-product of pigments and dyes create a regulatory burden for paper recycling, municipal treatment works, and other dischargers in Washington State. Washington standards to protect human health under the Clean Water Act (National Toxics Rule, 40 CFR 131.36) allow only 0.00000017 ppm (170 ppq) PCBs in surface water bodies, almost 300 million times lower than the allowable level of inadvertent PCBs in commercial products (50 ppm under TSCA). There are permitted releases in Washington State (e.g., paper mills that recycle materials and municipal wastewater treatment plants) that transport PCBs from sources (e.g., pigments in paper and other consumer goods) into waterways. Therefore, as a result of permitted releases, some PCBs from pigments are released into the environment, even though the permittees are not the source of PCBs. For example, PCB-11 and PCB-209 have been found in Washington State’s environment (EIM Database 2014). It is suspected that dyes from clothing and other consumer products (e.g., soaps, lotions, and creams) are also contributing PCBs to municipal wastewater treatment plants. Using alternatives assessment to find safer alternatives may reduce PCB

releases to the environment and alleviate water treatment burdens on businesses and municipalities.

PCBs have been detected in general consumer products, purchased in Washington State (Ecology 2014). Four congeners, known to be associated with pigments (i.e., PCB 11, 206, 208, and 209), were analyzed in 74 samples from 68 products. Products included packaging, paper materials, paint, colorants, and caulk. PCB-11 was detected in 49 products across all product categories at concentrations ranging from 1 to 48.5 ppb. PCB-209 was detected in one paper material and six paints or colorants up to 320 ppb. The Washington Dept. of Ecology (Ecology) is in the process of reporting on the results of all the congeners for the same samples, as well as additional samples. As in previously published work (Hu and Hornbuckle 2010), we also observed a broad distribution of congeners in consumer products.

Recommendations

Based on PCB sources in anthropogenic materials in Washington State (Table 1), as well as PCBs in environmental reservoirs (e.g., soil, sediment), we recommend eight new actions to complement existing state programs to reduce exposures to PCBs.

1. Survey and assess PCB-containing lamp ballasts in schools and other public buildings. Encourage replacement with more energy efficient PCB-free fixtures. This will remove aging lamp ballasts before they fail and expose children and staff while also improving energy efficiency. We are particularly concerned about children's exposure because they are more sensitive to harmful effects (Stone and Delistraty 2010). It is uncertain how many pre-1979 lamp ballasts are still in use, but these ballasts should be removed and/or replaced to reduce potential human exposure and increase energy efficiency. PCB-containing lamp ballasts are potentially in many older buildings, but the state should focus on buildings that are publically owned. There have been grants available from the Washington State Office of Superintendent of Public Instruction (OSPI) for schools for energy efficiency that can be used to replace the aging ballasts. OSPI has not tracked how many schools have already used the grants to replace PCB-containing lamp ballasts but can do so in the future.
2. Develop and promote Best Management Practices (BMPs) for containment of PCBs in buildings currently in use and those slated for demolition. A large quantity of PCBs in old caulk and other building materials is slowly being released into the environment. PCB releases from building materials can be greatly accelerated during remodeling and demolition. There is some guidance (e.g., EPA) on removing PCB-containing building materials.
3. Assess schools and other public buildings for the presence of PCB-containing building materials (e.g., caulk, paint). If these materials were removed, this would further reduce PCBs in schools. Focusing on children in schools should be a priority because children are more sensitive to PCBs, and schools are publically owned. However, other public buildings should also be assessed. Based on dates of construction and renovation of school buildings that were identified as part of Recommendation no. 1, schools should be prioritized for testing and remediation, pending on the availability of funds. Ecology should work with OSPI and other state agencies to incorporate requirements for PCB testing and BMPs into contracts for remediation and renovation of school buildings and other publically owned buildings.
4. Learn more about what products contain PCBs and promote the use of processes that do not inadvertently generate PCBs. Information on PCBs in products is important to implement Washington's new state purchasing law (RCW 39.26.280 2014) that directs state agencies to avoid purchasing products or product packaging that contains PCBs above the practical quantification limit (PQL), unless it is neither cost-effective nor technically feasible. Ecology has a product testing program that includes testing products to comply with this new law and has also collaborated with seven other states to develop guidance for alternatives assessment (IC2 2013). Future work should both determine which processes are likely to generate PCBs and continue product testing to identify products that contain PCBs. One source of potential processes comes from the rulemaking on inadvertent generation of PCBs. EPA generated a list of 200 chemical processes with a potential for generating PCBs and narrowed this to 70 processes with a high potential to inadvertently generate PCBs. To date, there has been no further work by EPA on processes that actually generate PCBs.
5. Survey owners of historic electrical equipment. This is to confirm what electrical equipment has been replaced and to provide technical assistance for proper replacement and disposal of the remaining equipment. While about 75 % of PCBs produced prior to 1979 were used in electrical equipment (EPA 1997), most transformers and large capacitors in Washington State that contained PCBs have been identified and replaced. The remaining equipment is generally monitored for spills that are cleaned up, further reducing the impact to humans and the environment. Federal TSCA regulations require proper use and disposal of identified PCB-contaminated items but do not require testing to

identify sources, so many unauthorized uses are not discovered until a release to the environment has occurred.

6. Expand environmental monitoring to identify any new locations requiring cleanup and determine the extent and relative importance of the air deposition pathway. Remedial technologies offer alternatives for reducing PCBs.
7. Conduct a public educational campaign to provide information to state residents on methods to minimize exposure to PCBs (e.g., lamp ballasts, small transformers in appliances).
8. Conduct a study to identify specific PCB congeners present in tissues of Washington State residents. This would clarify the extent of human exposure and the relative contribution of distinct congeners. This is particularly important in Washington State, given the elevated levels of PCBs in local fish populations and our relatively high fish consumption rate.

Conclusion

Although PCBs were banned in the USA in 1979, they are still ubiquitously distributed in the environment and produce multiple adverse effects in humans and wildlife. Current PCB levels in the environment represent both historical and ongoing loadings of PCBs in anthropogenic materials, as well as PCBs contained in environmental media. We evaluated PCBs in anthropogenic materials in Washington State and found the mass estimates much higher compared to environmental compartments. While the largest use of PCBs produced before 1979 was for transformers and large capacitors, we found that most transformers and large capacitors have been disposed of. Our recommendations to further reduce and phase out uses, releases, and exposures of PCBs focus on other sources, namely pre-1979 lamp ballasts and caulk, especially in schools, and current inadvertent generation. Small amounts from numerous dispersed sources with low levels of PCBs add up to levels that are of concern to human health and wildlife. Continued environmental monitoring, biomonitoring, cleanup, and education are also needed. Chemical Action Plans are useful to address the special concerns and challenges of PBTs, where traditional single-media approaches are not sufficient. Our findings and recommendations in Washington State are relevant to other geographic areas, both within the USA and globally, as the use patterns and reduction strategies have been similar among states and countries.

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Electronic Supplementary Material submitted to Environmental Science and Pollution Research for the following article;

Evaluation of PCB Sources and Releases for Identifying Priorities to Reduce PCBs in Washington State (USA)

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Electronic Supplementary Material includes 2 tables named, “Online Resource 1” and “Online Resource 2”

Online Resource 1. Registered transformers in Washington State. We attempted to contact the registrants to identify the current status of the registered transformers. While owners of PCB transformers (>500 ppm PCBs) were required to register with the EPA, EPA is not required to update the database.

Company			Transformer					
Name	City	Contact	Street address	City	Zip code	Number	Weight (kg)	Current status
Puget Sound Energy	Bellevue	John Rork	Talcott Ave & Columbia Street	Olympia	98501	3	7	In use
Puget Sound Energy	Bellevue	John Rork	14401 278th Ave NE	Duval	98019	2	5	In use
Puget Sound Energy	Bellevue	Lea Boyle	14401 188th Ave NE	Redmond	98052	2	4.52	In use
Puget Sound Energy	Bellevue	John Rork	S 173rd & 43rd Ave S	Renton	98055	0	0	
Puget Sound Energy	Bellevue	John Rork	2211 Nevada St	Bellingham	98225	0	0	
Puget Sound Energy	Bellevue	John Rork	24810 156th Ave SE	Kent	98025	0	0	
Puget Sound Energy	Bellevue	John Rork	Hodgedon & Garfield St	Tenino	98589	0	0	
Puget Sound Energy	Bellevue	John Rork	70th Street E & Myers Rd	Bonney Lk	98390	0	0	
Puget Sound Energy	Bellevue	John Rork	Dolarway Rd	Ellensburg	98922	0	0	
Puget Sound Energy	Bellevue	John Rork	Jackson & Main St	Cle Elum	98922	0	0	
Puget Sound Energy	Bellevue	John Rork	19319 Electron Rd	Orting	98360	0	0	
Puget Sound Energy	Bellevue	John Rork	W side of Stottlemeyer Rd	Poulsbo	98370	0	0	
Puget Sound Energy	Bellevue	John Rork	40801 268th Ave SE	Enumclaw	98022	0	0	
Puget Sound Energy	Bellevue	John Rork	N. Tapps Highway & Vandermark Rd	Auburn	98002	0	0	
Puget Sound Energy	Bellevue	John Rork	SE 80th St & 246 Ave SE	Issaquah	98027	0	0	
Puget Sound Energy	Bellevue	John Rork	13635 SE 26th	Bellevue	98004	0	0	
Puget Sound Energy	Bellevue	John Rork	3975 E. Highway 525	Langley	98260	0	0	
Puget Sound Energy	Bellevue	John Rork	1274 Thompson Rd	Anacortes	98221	0	0	
Puget Sound Energy	Bellevue	John Rork	2857 S. 221st	Des Moines	98148	0	0	
Puget Sound Energy	Bellevue	John Rork	12251 Mt Baker Highway	Glacier	98244	0	0	
Puget Sound Energy	Bellevue	John Rork	7537 Portal Way	Ferndale	98248	0	0	

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Company			Transformer					
Name	City	Contact	Street address	City	Zip code	Number	Weight (kg)	Current status
Puget Sound Energy	Bellevue	John Rork	13635 NE 80th	Redmond	98052	0	0	
Puget Sound Energy	Bellevue	John Rork	9512 Pacific Highway SE	Lacey	98503	0	0	
Puget Sound Energy	Bellevue	John Rork	9221 Wilows Rd NE	Redmond	98502	0	0	
Puget Sound Energy	Bellevue	John Rork	34717 21st Ave SW	Federal Way	98003	0	0	
Puget Sound Energy	Bellevue	John Rork	1035 Stevenson Ave	Enumclaw	98022	0	0	
Puget Sound Energy	Bellevue	John Rork	Hanford Road & Centralia Steam Plt	Centralia	98531	0	0	
Puget Sound Energy	Bellevue	John Rork	S of I-90 between Exits 37 & 38	Snoqualmie	98065	0	0	
Puget Sound Energy	Bellevue	John Rork	Corner of Central Valley Rd & Bucklin	Bremerton	98310	0	0	
Puget Sound Energy	Bellevue	John Rork	20th St E & 169th Ave E (2111)	Sumner	98340	0	0	
Western Washington University	Bellingham	Gayle Shipley	Commissary 781 25th St	Bellingham	98225	0	0	
SDS Lumber Co	Bingen	Ronald Schultz	South Side BNSF RR	Bingen	98605	2	2138	Unknown
Kimberly-Clark Worldwide	Everett	Jim Ketchum	2600 Federal Ave	Everett	98201	0	0	
Grays Harbor Paper L.P.	Hoquiam	Richard Johnston	801 23rd St	Hoquiam	98550	5	50,932	In use
Reynolds Metals Company	Longview	H.S. Hays	4029 Industrial Way	Longview	98632	0	0	
Washington Veneer	Omak	Joe Atwood	1100 Eighth Ave E	Omak	98841	7	12,412	Unknown
PUD. No. 1 of Clallam Co	Port Angeles	Quimby Moon	1936 W 18th St	Port Angeles	98362	4	505	Disposed of
City of Port Angeles	Port Angeles	Mark Shamp	321 E Fifth St	Port Angeles	98362	1		Disposed of
PUD. No. 1 of Clallam Co	Port Angeles	Quimby Moon	1936 W 18th St	Port Angeles	98363	1	100	Disposed of
PUD No. 1 of Clallam Co	Port Angeles	Quimby Moon	1936 W 18th St	Port Angeles	98363	1	68	Disposed of

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Company			Transformer					
Name	City	Contact	Street address	City	Zip code	Number	Weight (kg)	Current status
Port Townsend Paper Corporation	Port Townsend	John M. Recht	100 Mill Hill Rd	Port Townsend	98368	0	0	
City of Richland	Richland	Wayne Collop	806 Thayer Dr	Richland	99352	2	45	Disposed of
US Dept of Energy Richland Oper. Office	Richland	B.J. Dixon	200 E Area	Richland	99352	1	137	Unknown
Energy Northwest	Richland	J.P. Chasse	HPCS Diesel Generator Rm, Nuclear Plant #2, N Power Plant Loop	Blank	Blank	0	0	
Entercom Communications Corp	Seattle	Martin Hadfield	910 Lone Oak Rd	Longview	Blank	0	0	
Total Reclaim, Inc	Seattle	Craig Lorch	2200 Sixth Ave S	Seattle	98134	1	215	Disposed of
Seattle City Light	Seattle	Karen Dinehart	Laurelhurst Lane and 51st	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	4502 NE 41st St	Seattle	98124	3	182	Disposed of
Seattle City Light	Seattle	Karen Dinehart	Bellevue Ave E & E John	Seattle	98124	3	160	Disposed of
Seattle City Light	Seattle	Karen Dinehart	2826 NW Market St	Seattle	98124	2	114	Disposed of
Seattle City Light	Seattle	Karen Dinehart	7710 35th Ave. SW	Seattle	98124	1	68	Disposed of
Seattle City Light	Seattle	Karen Dinehart	6730 24th Ave. NW	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	1414 NW Leary Way	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	7750 28th Ave NW	Seattle	98124	0	0	
Seattle City Light	Seattle	Karen Dinehart	1405 NW 65th St	Seattle	98124	0	0	

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Company			Transformer						
Name	City	Contact	Street address	City	Zip code	Number	Weight (kg)	Current status	
Seattle City Light	Seattle	Karen Dinehart	8032 15th Ave NW	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	3209 NW 65th St	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	2333 W Boston St	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	2100 SW Andover St	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	35th Ave SE & SW Genessee	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	5601 23rd Ave SW	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	1605 SW Holden St	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	3405 SW Graham St	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	4118 SW Morgan St	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	9370 52nd Ave S	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	1st E/of Earl Ave NW, S/SI NW 90th	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	1st N/of S Holden, E/SI Rainier Ave S	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	1st S/of W Bertona, E/SI 21st Ave W	Seattle	98124	0	0		
Seattle City Light	Seattle	Karen Dinehart	48th NE & 47th NE	Seattle	98124	0	0		

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Company			Transformer					
Name	City	Contact	Street address	City	Zip code	Number	Weight (kg)	Current status
Seattle City Light	Seattle	Karen Dinehart	51st Ave NE & NE 41st St	Seattle	98124	0	0	
Inland Power and Light	Spokane	Todd Hoffman	10110 W Hallett Rd	Spokane	99014	0	1,249	
Avista Utilities	Spokane	Clarice Robertson	various locations	Blank	Blank	157	16,434	Disposed of
Avista Corporation	Spokane	Clarice Robertson	Onion Creek Rd	Colville	Blank	0	0	
Avista Corporation	Spokane	Clarice Robertson	SE corner of Rockwell and Monroe Sts	Spokane	Blank	0	0	
Tacoma Power	Tacoma	Russell Post	418 Gershick Rd	Silver Creek	98585	10	830	Disposed of
Tacoma School District #10	Tacoma	Margaret Ohlson	111 N E St	Tacoma	98403	1	358	In use
Tacoma School District #10	Tacoma	Margaret Ohlson	2502 N Orchard	Tacoma	98406	1	358	In use
Pioneer Americas, Inc./Chlor Alkali Co. Inc.	Tacoma	Karl Iams	605 Alexander Ave	Tacoma	98421	0	0	
TransAlta of Calgary	Alberta	Roger Carter	913 Big Hanaford Rd	Centralia	98531	42	34,731	Disposed of

Online Resource 2. Reports to EPA on inadvertent generation of PCBs (1994-present). Manufacturers are required to report inadvertent generation of PCBs to EPA.

Date	Reporter Company	Product	PCB concentration or amount	Category
4/13/1995	Sun Chem. Corp	2-Naphthalenecarboxylic acid, 4-[(2,5-dichlorophenyl) azo]-3-hydroxy, a dye precursor		pigments and dyes
2/11/2004	Clariant	imported dyes		pigments and dyes
6/13/2005	Clariant	imported dyes		pigments and dyes
5/19/2011	Clariant	imported dyes		pigments and dyes
7/29/1994	Ciba-Geigy Pigments Division	CBI ^a		pigments and dyes
12/28/1994	Ciba-Geigy Pigments Division	CBI		pigments and dyes
12/29/1994	DIC Trading	3 pigments		pigments and dyes
6/22/1995	Ciba-Geigy Pigments Division	CBI		pigments and dyes
7/25/1995	Cappelle	4 pigments		pigments and dyes
7/2/1996	Uhlich Color Co	CI Pigment Orange 24		pigments and dyes
7/15/1996	Ciba-Geigy Pigments Division	CBI		pigments and dyes
8/16/1996	Engelhard	CI Pigment Violet 23	19.6 ppm	pigments and dyes
8/23/1996	Cappelle	CI Pigment Yellow 170		pigments and dyes
9/27/1996	UMC (United Mineral and Chem)	CI Pigment Green 7		pigments and dyes
1/13/1997	Zeneca	7 pigments		pigments and dyes
7/29/1996	CDR Pigments and Dispersions	6 pigments		pigments and dyes
6/18/1997	Fabricolor	12 pigments		pigments and dyes
7/1/1997	BASF	13 pigments		pigments and dyes
8/18/1997	Ciba Pigments Division	CBI for several pages		pigments and dyes
10/21/1997	Mil International	5 pigments		pigments and dyes
1/6/1998	Sun Chem. Corp	4 pigments		pigments and dyes
10/26/1997	Mil International	4 pigments		pigments and dyes

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Date	Reporter Company	Product	PCB concentration or amount	Category
5/15/1998	Mil International	5 pigments		pigments and dyes
7/20/1998	Ciba Pigments Division	CBI		pigments and dyes
10/23/1998	Ciba Pigments Division	CBI		pigments and dyes
2/2/1999	Lansco Colors	7 pigments		pigments and dyes
7/15/1999	Ciba Colors Division	CBI		pigments and dyes
7/31/1999	Sun Chem. Corp	CBI		pigments and dyes
2/2/2000	Ciba Colors Division	CBI		pigments and dyes
5/23/2000	Ciba Colors Division	CBI		pigments and dyes
8/31/2000	Ciba Colors Division	CBI		pigments and dyes
9/8/2000	Avecia	7 pigments		pigments and dyes
11/22/2000	Mil International	7 pigments		pigments and dyes
12/13/2000	Ciba Colors Division	CBI		pigments and dyes
3/30/2001	Ciba Colors Division	CBI		pigments and dyes
5/4/2001	Magruder Color Co	3 pigments		pigments and dyes
6/1/2001	Sun Chem. Corp	9 pigments		pigments and dyes
7/18/2001	Ciba Colors Division	CBI		pigments and dyes
4/8/1994	PCL Group	Copper Phthalocyanine Blue		pigments and dyes
10/17/2001	Ciba Colors Division	CBI		pigments and dyes
1/25/2002	Ciba Colors Division	CBI		pigments and dyes
3/27/2002	Mil International	8 pigments		pigments and dyes
4/29/2002	Ciba Coating Effects	CBI		pigments and dyes
8/6/2002	Ciba Coating Effects	CBI		pigments and dyes
8/28/2002	Sun Chem. Corp	CBI		pigments and dyes

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Date	Reporter Company	Product	PCB concentration or amount	Category
11/5/2002	Ciba Coating Effects	CBI		pigments and dyes
6/13/2003	Ciba Coating Effects	CBI		pigments and dyes
10/16/2003	Ciba Coating Effects	CBI		pigments and dyes
4/2/2004	Ciba Coating Effects	CBI		pigments and dyes
7/6/2004	Ciba Coating Effects	CBI		pigments and dyes
7/6/2004	Ciba Coating Effects	CBI		pigments and dyes
8/8/2005	Sun Chemical	CBI		pigments and dyes
5/25/2006	Cappelle	CI Pigment Yellow 17		pigments and dyes
1/30/1995	GE Silicones	CBI	<2.5 ppm discounted, total discounted quantity <1.1 lbs	silicones
1/30/1996	GE Silicones	CBI	<1.1 ppm discounted, total discounted quantity <0.9 lbs	silicones
1/24/1997	GE Silicones	CBI	<1.5 ppm discounted, total discounted quantity <0.6 lbs	silicones
1/24/1997	GE Silicones	CBI	<1.3 ppm discounted, total discounted quantity <0.53 lbs	silicones
2/25/1999	GE Silicones	CBI	<1.5 ppm discounted, total discounted quantity <0.8 lbs	silicones
2/7/2000	GE Silicones	CBI	<1.7 ppm discounted, total discounted quantity <0.5 lbs	silicones
3/13/2001	GE	CBI	<1.9 ppm discounted, total discounted quantity <0.7 lbs	silicones
5/28/2002	GE	CBI, adding hydrolyzed phenylchlorosilanes and phenylchlorosilanes	total discounted quantity < 0.83 lbs	silicones
4/30/1997	ABB	electrical capacitors	3.9 ppm, 134 liters	Unique
6/24/1994	Nagase America	2,4,6-TCPH (2,4,6-Trichlorophenylhydrazine)	9-12 ppm	Unique
11/30/1995	PHT International	2,6-Dichloro-4-Nitro Aniline		Unique

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Date	Reporter Company	Product	PCB concentration or amount	Category
3/17/1998	ISK Biosciences	CBI, Chlorothalonil production		Unique
5/15/2001	PPG Industries	trichlorobenzene (TCB)		Unique
8/17/2012	Future Fuel	pesticide intermediate		Unique
4/7/1997	Elf Atochem		4 and 5 ppm	Unknown
2/18/2000	CBI	CBI		Unknown
6/13/2001	CBI	CBI		Unknown
2/4/2003	CBI	CBI		Unknown
CBI	CBI	CBI		Unknown
5/31/2011	CBI	CBI		Unknown
9/11/2012	CBI	CBI, 220 kg shipment		Unknown
8/23/2004	Formosa Plastics		up to 215-255 ppm, 143 lbs	vinyl chloride
6/24/1996	Geon		740 lbs PCB/62,676,000 lbs chemical feedstocks	vinyl chloride
11/13/1997	Dow			vinyl chloride

*CBI=Confidential Business Information.